

| | | | |
|--|--|---|--|
| FORM PTO-1300 (Modified) REV. 11-98 | | U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE | |
| TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371 | | ATTORNEY'S DOCKET NUMBER JMYT-246US | |
| INTERNATIONAL APPLICATION NO. PCT/GB00/00390 | | U.S. APPLICATION NO. (IF KNOWN, SEE 37 CFR To Be Assigned 09/913433 | |
| | | PRIORITY DATE CLAIMED 15 February 1999 (15.02.99) | |
| TITLE OF INVENTION NONWOVEN WEB | | | |
| APPLICANT(S) FOR DO/EO/US GASCOYNE, John Malcolm; RALPH, Thomas Robertson; and BROWN, Karen Leanne | | | |
| Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information: | | | |
| <ol style="list-style-type: none"> <input checked="" type="checkbox"/> This is a FIRST submission of items concerning a filing under 35 U.S.C. 371. <input type="checkbox"/> This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. 371. <input checked="" type="checkbox"/> This is an express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1). <input checked="" type="checkbox"/> A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date. <input checked="" type="checkbox"/> A copy of the International Application as filed (35 U.S.C. 371 (c) (2)) <ol style="list-style-type: none"> <input checked="" type="checkbox"/> is transmitted herewith (required only if not transmitted by the International Bureau). <input type="checkbox"/> has been transmitted by the International Bureau. <input type="checkbox"/> is not required, as the application was filed in the United States Receiving Office (RO/US). <input type="checkbox"/> A translation of the International Application into English (35 U.S.C. 371(c)(2)). <input checked="" type="checkbox"/> A copy of the International Search Report (PCT/ISA/210). <input checked="" type="checkbox"/> Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371 (c)(3)) <ol style="list-style-type: none"> <input type="checkbox"/> are transmitted herewith (required only if not transmitted by the International Bureau). <input type="checkbox"/> have been transmitted by the International Bureau. <input type="checkbox"/> have not been made; however, the time limit for making such amendments has NOT expired. <input checked="" type="checkbox"/> have not been made and will not be made. <input type="checkbox"/> A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)). <input checked="" type="checkbox"/> An oath or declaration of the inventor(s) (35 U.S.C. 371 (c)(4)). <input checked="" type="checkbox"/> A copy of the International Preliminary Examination Report (PCT/IPEA/409). <input type="checkbox"/> A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371 (c)(5)). | | | |
| Items 13 to 20 below concern document(s) or information included: | | | |
| <ol style="list-style-type: none"> <input type="checkbox"/> An Information Disclosure Statement under 37 CFR 1.97 and 1.98. <input checked="" type="checkbox"/> An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included. <input checked="" type="checkbox"/> A FIRST preliminary amendment. <input type="checkbox"/> A SECOND or SUBSEQUENT preliminary amendment. <input type="checkbox"/> A substitute specification. <input type="checkbox"/> A change of power of attorney and/or address letter. <input checked="" type="checkbox"/> Certificate of Mailing by Express Mail <input type="checkbox"/> Other items or information: <div style="border: 1px solid black; height: 100px; width: 100%;"></div> | | | |

U.S. APPLICATION NO (IF KNOWN, SEE 37 CFR
INTERNATIONAL APPLICATION NO

09/913453

PCT/GB00/00390

ATTORNEY'S DOCKET NUMBER

JMYT-246US

21. The following fees are submitted:

BASIC NATIONAL FEE (37 CFR 1.492 (a) (1) - (5)) :

| | |
|---|----------|
| <input type="checkbox"/> Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2) paid to USPTO and International Search Report not prepared by the EPO or JPO | \$970.00 |
| <input type="checkbox"/> International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO | \$840.00 |
| <input type="checkbox"/> International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO | \$690.00 |
| <input type="checkbox"/> International preliminary examination fee paid to USPTO (37 CFR 1.482) but all claims did not satisfy provisions of PCT Article 33(1)-(4) | \$670.00 |
| <input type="checkbox"/> International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims satisfied provisions of PCT Article 33(1)-(4) | \$96.00 |

ENTER APPROPRIATE BASIC FEE AMOUNT =

\$860.00

Surcharge of \$130.00 for furnishing the oath or declaration later than 20 30 months from the earliest claimed priority date (37 CFR 1.492 (e)).

\$0.00

| CLAIMS | NUMBER FILED | NUMBER EXTRA | RATE | |
|--|--------------|--------------|-----------|----------|
| Total claims | 45 - 20 = | 25 | x \$18.00 | \$450.00 |
| Independent claims | 3 - 3 = | 0 | x \$80.00 | \$0.00 |
| Multiple Dependent Claims (check if applicable). | | | ☒ | \$270.00 |

TOTAL OF ABOVE CALCULATIONS =

\$1,580.00

Reduction of 1/2 for filing by small entity, if applicable. Verified Small Entity Statement must also be filed (Note 37 CFR 1.9, 1.27, 1.28) (check if applicable).

\$0.00

SUBTOTAL =

\$1,580.00

Processing fee of \$130.00 for furnishing the English translation later than 20 30 months from the earliest claimed priority date (37 CFR 1.492 (f)).

\$0.00

TOTAL NATIONAL FEE =

\$1,580.00

Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31) (check if applicable).

☒ \$40.00

TOTAL FEES ENCLOSED =

\$1,620.00

| | |
|---------------------------|----|
| Amount to be: refunded | \$ |
| charged | \$ |

- A check in the amount of **\$1,620.00** to cover the above fees is enclosed.
- Please charge my Deposit Account No. **18-0350** in the amount of **A duplicate copy of this sheet is enclosed.** to cover the above fees.
- The Commissioner is hereby authorized to charge any fees which may be required, or credit any overpayment to Deposit Account No. **18-0350** A duplicate copy of this sheet is enclosed.

NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO:

Christopher R. Lewis
RATNER & PRESTIA
Suite 301
One Westlakes, Berwyn
P.O. Box 980
Valley Forge, PA 19482-0980

Telephone (610) 407-0700
Facsimile (610) 407-0701

SIGNATURE

Christopher R. Lewis

NAME

36,201

REGISTRATION NUMBER

August 14, 2001

DATE

JMYT-246US

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant: Karen Leanne Brown et al. : Art Unit:
Application No.: To Be Assigned : Examiner:
Filed: Herewith :
FOR: NONWOVEN WEB :

PRELIMINARY AMENDMENT

Assistant Commissioner for Patents
Washington, DC 20231

SIR:

Prior to examination, please amend the above-identified application as follows.

IN THE SPECIFICATION

Please replace the paragraph beginning at page 5, line 26, with the following paragraph:

The non-woven web may be made by a single individual process or by adapting a continuous manufacturing process, such as paper making. In both cases the fibres (long and short) are dispersed in solution, preferably water, to form a slurry. Also added to the slurry are one or more polymers (the "first polymer"), preferably hydrophilic polymers, for example polyvinylalcohol (PVA). The first polymer may be in the form of fibres. Once the long and short fibres and the first polymer are uniformly dispersed in the liquid, the liquid is either drained in the case of a single individual process or the fibres are formed into a continuous structure by the controlled deposition of the slurry onto a moving bed mesh. In such a wet laid process, the longer fibres, which exceed the thickness of the final structure typically by at least an order of magnitude, are pulled down by the water flow during the filtration process to lie in the x-y plane. Because of the density of

fibres, as each layer deposits, the longer fibres span across a number of other fibres, thus generally maintaining the x-y orientation. The short fibres, being generally of a similar length or shorter than the thickness of the final web, tend to be pulled by the water flow into the interstices of the web, formed by the longer fibres, but do not tend to span more than one of the longer fibres. Thus there is much greater z-directionality of the shorter fibres. The web so-formed is dried in an oven, and if necessary placed in a solution of the final polymer, which may or may not be the same as the first polymer, allowed to dry and subsequently heat treated to set the final polymer, if used, or to set the first polymer. If it is not desirable for the first polymer to remain in the final web structure, it may be removed by this heat treatment or by an alternative appropriate process. In addition, any undesirable residues may be removed by the heat treatment or by an alternative appropriate process.

IN THE CLAIMS

Please replace claims 1-7, 9, 10, 12-14, 16-17 and 20-25 with the following amended claims:

1 1. (Amended) A non-woven fibre web comprising a plurality of
2 longer fibres in the x- and y-directions of average length greater than 5mm and a
3 plurality of shorter fibres of average length less than 3mm wherein at least a
4 proportion of the shorter fibres are orientated in the z-direction, and wherein the
5 plurality of shorter fibres is at least 20% of the total weight of fibres, and wherein
6 the density of the non-woven fibre web is from 0.1g/cm³ to 0.35g/cm³.

1 2. (Amended) A non-woven fibre web comprising up to and
2 including 80% by weight of longer fibres in the x- and y-directions of average
3 length greater than 5mm, and 20% or more by weight of shorter fibres of average
4 length less than 3mm wherein at least a proportion of the shorter fibres are
5 orientated in the z-direction, and wherein the density of the non-woven fibre web is
6 from 0.1g/cm³ to 0.35g/cm³.

1 3. (Amended) A non-woven fibre web according to claim 1
2 wherein the plurality of shorter fibres is no more than 85% by weight of total fibres
3 in the substrate.

1 4. (Amended) A non-woven fibre web according to claim 3
2 wherein the plurality of shorter fibres is no more than 70% by weight of the total
3 fibres.

1 5. (Amended) A non-woven fibre web according to claim 1 or 2
2 wherein the longer fibres have a maximum average length of 50mm.

1 6. (Amended) A non-woven fibre web according to claim 5
2 wherein the longer fibres are of average length of 5mm to 30mm.

1 7. (Amended) A non-woven fibre web according to claim 1 or 2
2 wherein the shorter fibres are of average length less than 2mm.

1 9. (Amended) A non-woven fibre web according to claim 1 or 2
2 wherein the shorter fibres have an average minimum length of 50 μm .

1 10. (Amended) A non-woven fibre web according to claim 1 or 2
2 wherein the longer fibres and shorter fibres are independently selected from the
3 group consisting of carbon, glass, silica, polymer, metal and ceramic fibres.

1 12. (Amended) A non-woven fibre web according to claim 1 or 2
2 wherein the density of the non-woven fibre web is from 0.1g/cm³ to 0.2g/cm³.

1 13. (Amended) A process for the preparation of a non-woven
2 fibre web according to claim 1, said process comprising the steps of:

3 (i) dispersing the longer and shorter fibres in solution to form a
4 slurry;

5 (ii) adding at least one polymer to the slurry;

14. (Amended) A gas diffusion substrate comprising a non-woven substrate comprising a non-woven fibre web as claimed in claim 1 or 2, and a filler material.

16. (Amended) A gas diffusion substrate according to claim 14 wherein the filler material comprises a catalyst material.

17. (Amended) A gas diffusion electrode comprising a gas diffusion substrate as claimed in claim 1 or 2, and an electrocatalyst material.

20. (Amended) A membrane electrode assembly comprising a gas diffusion electrode as claimed in claim 17, a second gas diffusion electrode and a solid polymer membrane.

21. (Amended) A membrane electrode assembly comprising a gas diffusion electrode as claimed in claim 17, a gas diffusion substrate, and a solid polymer membrane, wherein an electrocatalyst layer is applied to the side of the membrane facing the gas diffusion substrate.

22. (Amended) A membrane electrode assembly comprising a gas diffusion substrate as claimed in claim 14, a gas diffusion electrode, and a solid polymer membrane, wherein an electrocatalyst layer is applied to the side of the membrane facing the gas diffusion substrate.

1 23. (Amended) A membrane electrode assembly comprising a
2 gas diffusion substrate as claimed in claim 14, and a second gas diffusion substrate,
3 and a solid polymer membrane, wherein an electrocatalyst layer is applied to both
4 sides of the solid polymer membrane.

1 24. (Amended) A fuel cell comprising a gas diffusion substrate
2 as claimed in claim 14.

1 25. (Amended) A fuel cell comprising a gas diffusion electrode
2 as claimed in claim 17.

Respectfully submitted,



Christopher R. Lewis, Reg. No. 36,201
Attorney for Applicants

EEA/tmb

Dated: August 14, 2001

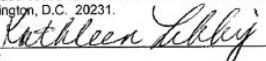
Suite 301
One Westlakes, Berwyn
P.O. Box 980
Valley Forge, PA 19482-0980
(610) 407-0700

The Assistant Commissioner for Patents is hereby
authorized to charge payment to Deposit Account
No. 18-0350 of any fees associated with this
communication.

EXPRESS MAIL Mailing Label No.: EL910364781US
Date of Deposit: August 14, 2001

I hereby certify that this paper and fee are being deposited,
under 37 C.F.R. § 1.10 and with sufficient postage, using the
"Express Mail Post Office to Addressee" service of the United
States Postal Service on the date indicated above and that
the deposit is addressed to the Assistant Commissioner for
Patents, Washington, D.C. 20231.

Kathleen Libby



VERSION WITH MARKINGS TO SHOW CHANGES MADEIN THE SPECIFICATION:

The non-woven web may be made by a single individual process or by adapting a continuous manufacturing process, such as paper making. In both cases the fibres (long and short) are dispersed in solution, preferably water, to form a slurry. Also added to the slurry are one or more polymers (the "first polymer"), preferably hydrophilic polymers, for example polyvinylalcohol (PVA). The first polymer may be in the form of fibres. Once the long and short fibres and the first polymer are uniformly dispersed in the liquid, the liquid is either drained in the case of a single individual process or the fibres are formed into a continuous structure by the controlled deposition of the slurry onto a moving bed mesh. In such a wet laid process, the longer fibres, which exceed the thickness of the final structure typically by at least an order of magnitude, are pulled down by the water flow during the filtration process to lie in the x-y plane. Because of the density of fibres, as each layer deposits, the longer fibres span across a number of other fibres, thus generally maintaining the x-y orientation. The short fibres, being generally of a similar length or shorter than the thickness of the final web, tend to be pulled by the water flow into the interstices of the web, formed by the longer fibres, but do not tend to span more than one of the longer fibres. Thus there is much greater z-directionality of the shorter fibres. The web so-formed is dried in an oven, and if necessary placed in a solution of the final polymer, which may or may not be the same as the first polymer, allowed to dry and subsequently heat treated to set the final polymer, if used, or to set the first polymer. If it is not desirable for the first polymer to remain in the final web structure, it may be removed by this heat treatment or by an alternative appropriate process. In addition, any undesirable residues may be removed by the heat treatment or by an alternative appropriate process.

IN THE CLAIMS:

1 1. (Amended) A non-woven fibre web comprising a plurality of
2 longer fibres in the x- and y-directions of average length greater than 5mm ("longer
3 fibres") and a plurality of shorter fibres of average length less than 3mm ("shorter
4 fibres") wherein at least a proportion of the shorter fibres of average length less
5 than 3mm are orientated in the z-direction, and wherein the proportion plurality of
6 shorter fibres of average length less than 3mm is at least 20% of the total weight of
7 fibres, and wherein the density of the non-woven fibre web is from 0.1g/cm³ to
8 0.35g/cm³.

1 2. (Amended) A non-woven fibre web comprising up to and
2 including 80% by weight of longer fibres in the x- and y-directions of average
3 length greater than 5mm ("longer fibres"), and 20% or more by weight of shorter
4 fibres of average length less than 3mm ("shorter fibres") wherein at least a
5 proportion of the shorter fibres of average length of less than 3mm are orientated in
6 the z-direction, and wherein the density of the non-woven fibre web is from
7 0.1g/cm³ to 0.35g/cm³.

1 3. (Amended) A non-woven fibre web according to claim 1 or
2 wherein the proportion plurality of shorter fibres is suitably no more than
3 85% by weight of total fibres in the substrate.

1 4. (Amended) A non-woven fibre web according to claim 3
2 wherein the proportion plurality of shorter fibres is suitably no more than 70% by
3 weight of the total fibres.

1 5. (Amended) A non-woven fibre web according to any of
2 claims 1 to 4 or 2 wherein the longer fibres suitably have a maximum
3 average length of 50mm.

1 6. (Amended) A non-woven fibre web according to claim 5
2 wherein the longer fibres are of average length of 5mm to 30mm.

1 7. (Amended) A non-woven fibre web according to any
2 preceding claim 1 or 2 wherein the shorter fibres are of average length less than
3 2mm.

1 9. (Amended) A non-woven fibre web according to any
2 preceding claim 1 or 2 wherein the shorter fibres have an average minimum length
3 of 50µm.

10. (Amended) A non-woven fibre web according to any
preceding claim 1 or 2 wherein the longer fibres and shorter fibres are
independently selected from the group consisting of carbon, glass, silica, polymer,
metal and ceramic fibres.

1 12. (Amended) A non-woven fibre web according to any
2 preceding claim 1 or 2 wherein the density of the non-woven fibre web is from
3 0.1g/cm³ to 0.2g/cm³.

13. (Amended) A process for the preparation of a non-woven
fibre web according to any preceding claim 1, said process comprising the steps of:

1 14. (Amended) A gas diffusion substrate comprising a non-
2 woven substrate comprising a non-woven fibre web as claimed in ~~any one of claims~~
3 ~~1 to 12~~ claim 1 or 2, and a filler material.

1 16. (Amended) A gas diffusion substrate according to claim 14
2 or ~~claim 15~~ wherein the filler material comprises a catalyst material.

1 17. (Amended) A gas diffusion electrode comprising a gas
2 diffusion substrate as claimed in ~~any one of claims 14, 15 and 16~~ claim 1 or 2, and
3 an electrocatalyst material.

1 20. (Amended) A membrane electrode assembly comprising a
2 gas diffusion electrode as claimed in ~~any one of claims 17, 18 and 19~~ claim 17, a
3 second gas diffusion electrode ~~which may or may not be an electrode as claimed in~~
4 ~~any one of claim 17, 18 and 19~~, and a solid polymer membrane.

1 21. (Amended) A membrane electrode assembly comprising a
2 gas diffusion electrode as claimed in ~~any one of claims 17, 18 and 19~~ claim 17, a
3 gas diffusion substrate ~~which may or may not be a substrate as claimed in any one~~
4 ~~of claims 14, 15 and 16~~, and a solid polymer membrane, wherein an electrocatalyst
5 layer is applied to the side of the membrane facing the gas diffusion substrate.

1 22. (Amended) A membrane electrode assembly comprising a
2 gas diffusion substrate as claimed in ~~any one of claims 14, 15 and 16~~ claim 14, a
3 gas diffusion electrode ~~which may or may not be an electrode as claimed in any one~~
4 ~~of claims 17, 18 and 19~~, and a solid polymer membrane, wherein an electrocatalyst
5 layer is applied to the side of the membrane facing the gas diffusion substrate.

1 23. (Amended) A membrane electrode assembly comprising a
2 gas diffusion substrate as claimed in ~~any one of 14, 15 and 16~~ claim 14, and a
3 second gas diffusion substrate ~~which may or may not be a substrate as claimed in~~
4 ~~any one of claims 14, 15 and 16~~, and a solid polymer membrane, wherein an
5 electrocatalyst layer is applied to both sides of the solid polymer membrane.

1 24. (Amended) A fuel cell comprising a gas diffusion substrate
2 as claimed in any one of claims 14, 15 and 16 claim 14.

1 25. (Amended) A fuel cell comprising a gas diffusion electrode
2 as claimed in any one of claims 17, 18 and 19 claim 17.

NONWOVEN WEB

5

The present invention relates to a novel porous substrate web a porous gas diffusion substrate and a porous gas diffusion electrode which may have application in electrochemical devices. for use for example in a fuel cell, and a process for the manufacture of the web, substrate and electrode.

10

Electrochemical cells invariably comprise at their fundamental level a solid or liquid electrolyte and two electrodes. the anode and cathode, at which the desired electrochemical reactions take place. Gas diffusion electrodes are employed in a range of electrochemical devices, in which a gaseous reactant and/or product has to be diffused into and/or out of one of the cell electrode structures. They are designed to optimise the contact between the reactant and the electrolyte to maximise the reaction rate. Electrocatalysts are often incorporated into gas diffusion electrode structures to increase the rates of the desired electrode reactions.

15

Gas diffusion electrodes are employed in many different electrochemical devices, including metal-air batteries, electrochemical gas sensors, electrosynthesis of useful chemical compounds, and in particular, fuel cells. Conventionally, gas diffusion electrodes comprise many components and are typically made up of one, two or even more layers of these components. Typically the gas diffusion electrode will comprise 20 one or more electrocatalyst containing layers, which are supported onto a more rigid porous substrate layer.

30

A fuel cell is an energy conversion device that efficiently converts the stored chemical energy of its fuel into electrical energy by combining either hydrogen, stored as a gas, or methanol stored as a liquid or gas, with oxygen to generate electrical power. The hydrogen or methanol are oxidised at the anode and oxygen is reduced at the cathode. Both electrodes are of the gas diffusion type. The electrolyte has to be in contact with both electrodes and may be acidic or alkaline, liquid or solid, in nature. In proton exchange membrane fuel cells (PEMFC), the electrolyte is a solid proton-conducting polymer membrane. commonly based on perfluorosulphonic acid materials,

and the combined structure formed from the membrane and the two gas diffusion electrodes is known as a membrane electrode assembly (MEA). Alternatively, the MEA may be formed from two porous gas diffusion substrates and a solid proton-conducting polymer membrane catalysed on both sides; or the MEA may be formed from one gas diffusion electrode and one gas diffusion substrate and a solid proton-conducting polymer catalysed on the side facing the gas diffusion substrate. The anode gas diffusion electrode or substrate is designed to be porous and to allow the reactant hydrogen or methanol to enter from the face of the electrode or substrate exposed to the reactant fuel supply, and then to diffuse through the thickness of the electrode or substrate to the reaction sites which contain electrocatalysts, usually platinum metal based, to maximise the electrochemical oxidation of hydrogen or methanol. The anode is also designed to allow electrolyte to penetrate through the face of the electrode or substrate exposed to the electrolyte and to also contact the same reaction sites. With acidic electrolyte types the product of the anode reaction are protons and these can then be efficiently transported from the anode reaction sites through the electrolyte to the cathode gas diffusion electrode or substrate. The cathode is also designed to be porous and to allow oxygen or air to enter the electrode or substrate and diffuse through to the reaction sites. Electrocatalysts are again commonly incorporated to maximise the rate of the reaction at the cathode reaction sites which combines the protons with oxygen to produce water.

Product water then has to diffuse out of the cathode structure. The structure of the cathode has to be designed such that it enables the efficient removal of the product water. If water builds up in the cathode, it becomes more difficult for the reactant oxygen to diffuse to the reaction sites, and thus the performance of the fuel cell decreases. In the case of methanol fuelled PEMFCs, additional water is present due to the water contained in the methanol, which can be transported through the membrane from the anode to the cathode side. The increased quantity of water at the cathode requires removal. However, it is also the case with proton conducting membrane electrolytes, that if too much water is removed from the cathode structure, the membrane can dry out resulting in a significant decrease in the performance of the fuel cell.

Traditionally, the gas porous substrates used in the PEMFC are based on high density materials such as rigid carbon fibre paper (i.e. Toray TGP-H-60 or TGP-H-90 from Toray Europe Ltd., 7 Old Park Lane, London, W1Y 4AD), with a bulk density of 0.49g/cm³ or woven carbon cloths, such as Zoltek PWB-3 (Zoltek Corporation, 3101 5 McKelvey Road, St. Louis, Missouri 63044) with a bulk density of 1.75g/cm³. Substrates such as these generally have good water management properties but either do not easily allow the passage of the reactant gases, in the case of the carbon fibre paper, or lack dimensional stability, as the cloth can easily be stretched in the directions of the major planar faces (x and y directions). In addition, these types of material are expensive 10 when compared to the material cost estimates needed to make fuels cells competitive with existing technologies, particularly in respect of mobile applications such as cars and buses.

More recently, electrode structures based on a porous substrate comprising a 15 non-woven network of carbon fibres, with a particulate material embedded within the fibre network as disclosed in EP-A-0 791 974 have shown comparable performances to structures based on carbon fibre paper or cloth, without the drawbacks of such materials, and at much lower fibre bulk densities, typically below 0.1g/cm³. Electrodes based on 20 non-woven carbon fibre structures such as Optimat® 203, of bulk density 0.07g/cm³ (from Technical Fibre Products, Kendal, Cumbria, UK) give a physically strong, dimensionally stable and handleable structure at a cost compatible with motive power applications. However, although the gas diffusion properties of such substrates are very 25 good, the structure lacks the ability to retain sufficient water, when operating at high current densities, to maintain optimum hydration of the membrane. As a consequence the resistance of the membrane increases with a commensurate loss in overall performance.

It is therefore an object of the present invention to provide a porous substrate, suitable for use for example in a gas diffusion electrode, which maintains the strength 30 and flexibility of the non-woven carbon fibre network substrate and retains the good gas diffusion properties, but also has greatly improved water management properties. Control of these properties is highly important to ensure the optimum functioning of the PEMFC

which may operate under a range of conditions of, for example, temperature, pressure, reactant gas flow rates and reactant gas level of humidification.

Accordingly, the present invention provides a non-woven fibre web comprising a plurality of fibres in the x- and y-directions of average length greater than 5mm and a plurality of fibres of average length less than 3mm wherein at least a proportion of the fibres of average length less than 3mm are orientated in the z-direction, and wherein the proportion of fibres of average length less than 3mm is at least 20% of the total weight of fibres, and wherein the density of the non-woven fibre web is from 0.1g/cm³ to 10 0.35g/cm³. In other words, the present invention provides a non-woven fibre web comprising up to and including 80% by weight of fibres in the x- and y-directions of average length greater than 5mm, and 20% or more by weight of fibres of average length less than 3mm wherein at least a proportion of the fibres of average length of less than 3mm are orientated in the z-direction, and wherein the density of the non-woven fibre 15 web is from 0.1g/cm³ to 0.35g/cm³.

The fibres of average length of greater than 5mm (the "longer fibres") impart physical strength to the web, whilst the fibres of average length of less than 3mm (the "shorter fibres") enable the density of the web to be increased beyond that which is 20 possible with longer fibres alone. As the proportion of shorter fibres increases so the strength of the web decreases and accordingly the proportion of shorter fibres is suitably no more than 85% by weight of total fibres in the substrate, preferably no more than 70% by weight of the total fibres.

25 The longer fibres will all be orientated in the x- and y-directions, while the shorter fibres may be orientated in the x-, y- and z-directions.

The longer fibres are of average length greater than 5mm and suitably have a maximum average length of 50mm. The preferred average length of the fibres is 5mm to 30mm. The diameter of the longer fibres is typically in the range of 0.2μm to 25μm, 30 preferably in the range of 2μm to 20μm.

The shorter fibres are of average length less than 3mm and suitably are of average length less than 2mm, preferably less than 1mm. The shorter fibres suitably have a minimum average length of 50 μm , preferably 100 μm . The diameter of the shorter fibres is typically in the range of 0.1 μm to 20 μm , preferably in the range of 0.4 μm to 10 μm .

- 5 Examples of such fibres include e.g. spun fibres and carbon fibre wools of type FRC 15 (ex Le Carbone (GB) Ltd) which are composed of mixed lengths generally between about 0.2mm and about 2mm.

Fibres which are suitable for use for the longer fibres and the shorter fibres include

- 10 carbon, glass, silica, polymer, metal or ceramic fibres, preferably carbon, silica, metal or ceramic, most preferably carbon. The fibres used to the longer and shorter fibres may be of the same material or different.

15 The density of the non-woven fibre web is suitably greater than 0.1g/cm³; suitably the maximum density of the non-woven fibre web is 0.35g/cm³. The preferred range is from 0.1g/cm³ to 0.2g/cm³.

The fibres in the web are held together by a polymer (the "final polymer"). Depending on the polymeric substance used it may also contribute to the essential 20 electrode structural properties in a gas diffusion substrate or electrode, such as control of the hydrophobic/hydrophilic balance. Examples of such polymers include polytetrafluoroethylene (PTFE), fluorinated ethylene-propylene (FEP), polyvinylidene difluoride (PVDF), Viton A, polyethylene, polypropylene, ethylene-propylene. The preferred final polymer is PTFE or FEP.

- 25 The non-woven web may be made by a single individual process or by adapting a continuous manufacturing process, such as paper making. In both cases the fibres (long and short) are dispersed in solution, preferably water, to form a slurry. Also added to the slurry are one or more polymers (the "first polymer"), preferably hydrophilic polymers, 30 for example polyvinylalcohol (PVA). The first polymer may be in the form of fibres.

Once the long and short fibres and the first polymer are uniformly dispersed in the liquid, the liquid is either drained in the case of a single individual process or the fibres are formed into a continuous structure by the controlled deposition of the slurry onto a moving bed mesh. The web so-formed is dried in an oven, and if necessary placed in a solution of the final polymer, which may or may not be the same as the first polymer, allowed to dry and subsequently heat treated to set the final polymer, if used, or to set the first polymer. If it is not desirable for the first polymer to remain in the final web structure, it may be removed by this heat treatment or by an alternative appropriate process. In addition, any undesirable residues may be removed by the heat treatment or
10 by an alternative appropriate process.

A second embodiment of the invention provides a gas diffusion substrate comprising a non-woven fibre web as hereinbefore defined and a filler material. The filler material is for the purpose of providing suitable gas diffusion, electrical conductivity and water management properties when employed as a gas diffusion substrate. Suitably, the filler material comprises a particulate carbon and a polymer, the carbon suitably being in the form of a powder. The carbon powder may be any of the materials generally designated as carbon black such as acetylene blacks, furnace blacks, pitch coke based powders and graphitised versions of such materials. Suitably also both natural and synthetic graphites may be used in this application. Such materials may be used either alone or in combination. The particulate carbon, or carbons, in the fill are held together by one or more polymers. The polymeric materials used will contribute to the essential electrode structural properties such as control of the hydrophobic/hydrophilic balance. Examples of such polymers include
15 polytetrafluoroethylene (PTFE), fluorinated ethylene-propylene (FEP), polyvinylidene difluoride (PVDF), Viton A, polyethylene, polypropylene, ethylene-propylene.
20 The preferred final polymer is PTFE or FEP.

The filler material may further comprise a catalyst other than an electrocatalyst, for example a gas phase catalyst, which is designed to remove contaminant gases in the fuel or oxidant feed streams as for example carbon monoxide in the hydrogen fuel, when
30 this is supplied from a reformer.

The most appropriate method for the manufacture of the gas diffusion substrate is to prepare the non-woven fibre web, for example using the method hereinbefore described, and subsequently in-filling with the filler material. The majority of the filler material will be forced into the structure of the non-woven fibre web, although a small quantity may remain on the surface. Alternatively, a continuous manufacturing process similar to that used to prepare the non-woven fibre web may be used, the filler material being added to the slurry.

A third aspect of the invention provides a gas diffusion electrode comprising a gas diffusion substrate as hereinbefore described and an electrocatalyst material. The electrocatalyst material is applied as a thin layer to the surface of the gas diffusion substrate. Some of the electrocatalyst material may penetrate slightly into the substrate, the remaining material forming a layer on the surface of the substrate. The electrocatalyst material comprises one or more electrocatalytic components and a polymer. Suitable polymers include hydrophobic polymers, such as PTFE and/or proton conducting polymers, such as Nafion® (ex DuPont). The electrocatalytic component is defined as a substance that promotes or enhances the rate of the electrochemical reaction of interest but remains unaltered by the reaction. The electrocatalytic component or components selected will depend on the application for which the gas diffusion electrode is being used. These may be, for example, a precious metal or a transition metal as the metal or metal oxide, either unsupported or supported in a dispersed form on a carbon support; a carbon or an organic complex, in the form of a high surface area finely divided powder or fibre, or a combination of these options. An example of a suitable electrocatalyst material is described in EP-A-0 731 520.

25

A fourth aspect of the invention provides a membrane electrode assembly comprising a gas diffusion electrode of the invention as hereinbefore defined and a second gas diffusion electrode which may or may not be an electrode according to the invention, and a solid polymer membrane, for example Nafion®. Alternatively, the invention provides a membrane electrode assembly comprising a gas diffusion electrode of the invention as hereinbefore defined, a gas diffusion substrate which may or may not be a substrate according to the invention and a solid polymer membrane, for example

Nafion®, wherein an electrocatalyst layer is applied to the side of the membrane facing the gas diffusion substrate. Alternatively, the invention provides a membrane electrode assembly comprising a gas diffusion substrate of the invention as hereinbefore defined, a gas diffusion electrode which may or may not be an electrode according to the invention and a solid polymer membrane, for example Nafion®, wherein an electrocatalyst layer is applied to the side of the membrane facing the gas diffusion substrate. Alternatively, the invention provides a membrane electrode assembly comprising a gas diffusion substrate of the invention as hereinbefore defined and a second gas diffusion substrate which may or may not be a substrate according to the invention, and a solid polymer membrane, for example Nafion®, wherein an electrocatalyst layer is applied to both sides of the solid polymer membrane.

A still further aspect of the invention provides a fuel cell comprising a gas diffusion substrate according to the present invention. A further aspect provides a fuel cell comprising a gas diffusion electrode according to the present invention.

The invention will now be further described with reference to the following Examples (according to the invention) and Comparative Example (not according to the invention).

The materials of the invention can be employed as either the anode or cathode, and indeed both anode and cathode in the electrochemical cells of the specific application. In the following examples, the electrodes are incorporated as the cathode in membrane electrode assemblies (MEAs) and evaluated in a proton exchange membrane fuel cell, with hydrogen as the anode fuel and air or pure oxygen as the cathode oxidant. It is at the cathode that the majority of cell performance (voltage) losses occur in cells operating with hydrogen as the fuel. The MEAs were fabricated by hot pressing the anode and cathode against each face of the solid proton conducting electrolyte membrane as is commonly practiced in the art.

The anodes were of the more conventional type, currently widely employed in the PEMFC. They comprised a convention pre-teflonated rigid conducting carbon fibre paper

substrate (Toray TGP-H-090, available from Toray Industries Inc., Tokyo, Japan) to which was applied a layer of a 20wt% platinum, 10wt% ruthenium catalyst, supported on Cabot's Vulcan® XC72R (from Johnson Matthey Inc., New Jersey, USA), at an electrode platinum loading of $0.25\text{mg}/\text{cm}^2$ of electrode geometric area. The MEAs were evaluated in a PEMFC single cell, with a geometric electrochemically active area of 240cm^2 . The single cell consisted of graphite plates into which flowfields were machined to distribute reactant gases and humidification water, and remove products. The MEA was located between the flow-field plates. The operation of the single cell was controlled from a purpose built test station facility. The "performance" of the fuel cell was assessed by measuring the voltage and current density relationship using a standard operating procedure. Unless otherwise stated, these conditions were typically, a reactant gas inlet temperature of 80°C , a pressure of both anode and cathode reactant gases of 3 atmosphere, and a reactant stoichiometry of 1.5 for hydrogen and 2.0 for air.

15

COMPARATIVE EXAMPLE 1

(not according to the invention)

A preformed non-woven carbon fibre structure was a $17\text{g}/\text{m}^2$ (equivalent to $0.07\text{g}/\text{cm}^3$) density carbon fibre paper (Optimat® 203, ex Technical Fibre Products, Kendal, Cumbria, UK). This was pre-coated with polytetrafluoroethylene (PTFE) by soaking the cloth for 1 minute in a solution of 20 parts by weight of PTFE emulsion (ICI's Fluon® dispersion GP1) in 500 parts by weight of water then draining and allowing to dry. The coated carbon fibre paper was heated to 350°C in air to sinter the PTFE.

25

The particulate material (the "filler material") used for embedding within the fibre network was provided by dispersing 47 weight parts of acetylene black (Shawinigan black from Chevron Chemicals, Houston, Texas, USA) in 1200 parts of water. To this was added 3 weight parts of PTFE as a dispersion in water (ICI's Fluon® dispersion GP1, 64 wt% solids suspension) and the mixture stirred to entrain the PTFE particles within the carbon black. The resultant material was dispersed using a high shear mixer (Silverson L4R) to produce a smooth mixture.

The particulate material was pressed into the non-woven carbon fibre structure from one side, and leveled off using a metal edge. The sheet was then dried at 200°C for 1 minute. A further thin layer of the particulate material was applied to the same side; the structure was sandwiched between two sheets of filter paper and passed through a set of rollers to compact the layer. The sheet was then dried at 200°C for 1 minute. This process was then repeated for the second side. Further additions of thin layers of the particulate material were applied to each side with compaction and drying until a loading of 3.33mg/cm² of carbon was achieved. The resulting gas diffusion substrate sheet was fired, in air, to 200°C for 30 minutes.

10

A catalyst material used for forming the electrocatalyst layer on the gas diffusion substrate was provided by dispersing 100 weight parts of a 40 weight % platinum on carbon black (Johnson Matthey FC40) in 30 parts of DuPont's Nafion® EW1100 as a 9.5 weight % dispersion in water, according to methods described in EP-A-0 731 520. The particulate catalyst was dispersed using a high shear mixer (Silverson L4R) to produce a smooth ink.

15

A layer of the catalyst material was then applied to the top face of the filled non-woven gas diffusion substrate, to give a loading of 0.71mg of platinum/cm². The electrode formed the cathode of an MEA, with the platinum electrocatalyst layer bonded to the membrane electrolyte face. The membrane employed was DuPont's Nafion® 112. The single cell results, on air and oxygen are shown in Figures 1 and 2 respectively.

20

EXAMPLE 1

1.0g of chopped carbon fibres at a fibre length of 6mm, and 1.0g of chopped carbon fibres at a fibre length of 12mm (type RK 10 supplied by RK Carbon Fibres Ltd.) along with 1.0g of carbon fibre wool composed of mixed lengths between 0.2mm and 2mm (type FRC 15 supplied by Le Carbone (Great Britain) Ltd., Portslade, Sussex, UK) and 0.2g of polyvinyl alcohol fibres (type Mewlon® SML supplied by Unitika Ltd., Oska 541, Japan) were dispersed in 3 litres of water using a standard catering blender. The resulting dispersion was used to prepare a sample of non-woven sheet of size 330mm diameter (855.3cm²) in a custom built sheet former (similar in general operation to a

standard SCA sheet former, as supplied by AB Lorentzen & Wettre, Box 4, S-163 93 Stockholm, Sweden). The sheet was dried at 100°C in air and had a density of 0.116g/cm³.

5 The non-woven carbon fibre sheet was placed on a sheet of PTFE (skived sheet from Dalau Ltd., Clacton-on-sea, Essex, UK) and sprayed with a suspension of PTFE 2.3g (ICI's Fluon® dispersion GP1, 64 wt% solids suspension) in 10g of water. Half of the suspension was sprayed on the first side, the sheet dried at 120°C, and the other side sprayed with the remaining suspension. The resulting sheet was fired, in air, to 340°C for
10 15 minutes, to give a 30% loading of PTFE.

The particulate material (prepared as described above in Comparative Example 1)
15 was pressed into the non-woven carbon fibre sheet from one side, and levelled off using a metal edge. The sheet was then dried at 200°C for 1 minute. A further thin layer of the particulate material was applied to the same side; the structure was sandwiched between two sheets of filter paper and passed through a set of rollers to compact the layer. The sheet was then dried at 200°C for 1 minute. This process was repeated for the second side. Further additions of thin layers of the particulate material were applied alternately to each side, with compaction and drying, until a loading of 3.33mg/cm² of
20 carbon was achieved. The resulting gas diffusion substrate sheet was fired, in air, to 300°C for 30 minutes.

A catalyst material used for forming the electrocatalyst layer on the gas diffusion substrate was provided as described above in Comparative Example 1.

25 A layer of the electrocatalyst material was then applied to the top face of the filled non-woven gas diffusion substrate, to give a loading of 0.8mg of platinum/cm².

30 The electrode formed the cathode of an MEA, with the platinum catalyst layer bonded to the membrane electrolyte face. The membrane employed was DuPont's Nafion® 112. The single cell results, on air and oxygen are shown in Figures 1 and 2 respectively.

EXAMPLE 2

1.0g of chopped carbon fibres at a fibre length of 6mm, and 1.0g of chopped carbon fibres at a fibre length of 12mm (type RK 10 supplied by RK Carbon Fibres Ltd.) along with 1.0g of carbon fibre wool composed of mixed lengths between 0.2mm and 2mm (Type Donacarbo MC232 supplied by Asland-Sudchemie-Kernfest GMBH, Hilden, Germany) and 0.3g of polyvinyl alcohol fibres (type Mewlon® SML supplied by Unitika Ltd., Oska 541, Japan) were dispersed in 3 litres of water using a standard catering blender. The resulting dispersion was used to prepare a sample of non-woven sheet of size 330mm diameter (855.3 cm^2) in a custom built sheet former (similar in general operation to a standard SCA sheet former, as supplied by AB Lorentzen & Wettre, Box 4, S-163 93 Stockholm, Sweden). The sheet was dried at 100°C in air and had a density of 0.13g/cm^3 .

15 The non-woven carbon fibre sheet was placed on a sheet of PTFE (skived sheet from Dalau Ltd., Clacton-on-sea, Essex, UK) and sprayed with a suspension of PTFE 2.3g (ICI's Fluon® dispersion GP1, 64 wt% solids suspension) in 10g of water. Half of the suspension was sprayed on the first side, the sheet dried at 120°C , and the other side sprayed with the remaining suspension. The resulting sheet was fired, in air, to 340°C for 15 minutes, to give a 30% loading of PTFE.

20 The particulate material (prepared as described above in Comparative Example 1) was pressed into the non-woven carbon fibre sheet from one side, and levelled off using a metal edge. The sheet was then dried at 200°C for 1 minute. A further thin layer of the particulate material was applied to the same side; the structure was sandwiched between two sheets of filter paper and passed through a set of rollers to compact the layer. The sheet was then dried at 200°C for 1 minute. This process was repeated for the second side. Further additions of thin layers of the particulate material were applied alternately to each side, with compaction and drying, until a loading of 6.03mg/cm^2 of carbon was achieved. The resulting gas diffusion substrate sheet was fired, in air, to 300°C for 30 minutes.

25 A catalyst material used for forming the electrocatalyst layer on the gas diffusion substrate was provided as described above in Comparative Example 1.

A layer of the electrocatalyst material was then applied to the top face of the filled non-woven gas diffusion substrate, to give a loading of 0.73mg of platinum/cm².

The electrode formed the cathode of an MEA, with the platinum catalyst layer bonded to the membrane electrolyte face. The membrane employed was DuPont's Nafion® 112. The single cell results, on air and oxygen are shown in Figures 1 and 2 respectively.

EXAMPLE 3

10 1.0g of chopped carbon fibres at a fibre length of 6mm, and 1.0g of chopped carbon fibres at a fibre length of 12mm (type RK 10 supplied by RK Carbon Fibres Ltd.) along with 1.0g of carbon fibre wool composed of mixed lengths between 0.2mm and 2mm (Type Donacarbo S232 supplied by Asland-Sudchemie-Kernfest GMBH, Hilden, Germany) and 0.25g of polyvinyl alcohol fibres (type Mewlon® SML supplied by Unitika Ltd., Oska 541, Japan) were dispersed in 3 litres of water using a standard catering blender. The resulting dispersion was used to prepare a sample of non-woven sheet of size 330mm diameter (855.3cm²) in a custom built sheet former (similar in general operation to a standard SCA sheet former, as supplied by AB Lorentzen & Wettre, Box 4, S-163 93 Stockholm, Sweden). The sheet was dried at 100°C in air and had a density of 0.12g/cm³.

15 The non-woven carbon fibre sheet was placed on a sheet of PTFE (skived sheet from Dalau Ltd., Clacton-on-sea, Essex, UK) and sprayed with a suspension of PTFE 2.3g (ICI's Fluon® dispersion GP1, 64 wt% solids suspension) in 10g of water. Half of the suspension was sprayed on the first side, the sheet dried at 120°C, and the other side sprayed with the remaining suspension. The resulting sheet was fired, in air, to 340°C for 15 minutes, to give a 30% loading of PTFE.

20 The particulate material (prepared as described above in Comparative Example 1) was pressed into the non-woven carbon fibre sheet from one side, and levelled off using a metal edge. The sheet was then dried at 200°C for 1 minute. A further thin layer of the particulate material was applied to the same side; the structure was sandwiched between two sheets of filter paper and passed through a set of rollers to compact the layer.

The sheet was then dried at 200°C for 1 minute. This process was repeated for the second side. Further additions of thin layers of the particulate material were applied alternately to each side, with compaction and drying, until a loading of 5.6mg/cm² of carbon was achieved. The resulting gas diffusion substrate sheet was fired, in air, to 5 300°C for 30 minutes.

A catalyst material used for forming the electrocatalyst layer on the gas diffusion substrate was provided as described above in Comparative Example 1.

10 A layer of the electrocatalyst material was then applied to the top face of the filled non-woven gas diffusion substrate, to give a loading of 0.71mg of platinum/cm².

15 The electrode formed the cathode of an MEA, with the platinum catalyst layer bonded to the membrane electrolyte face. The membrane employed was DuPont's Nafion® 112. The single cell results, on air and oxygen are shown in Figures 1 and 2 respectively.

20 The cell potential versus current density performance of the electrodes in Comparative Test 1 (not according to the invention) is typical of the performance of an electrode produced by filling conventional non-woven carbon structures. The gas diffusion properties of such substrates are very good, with the fall-off in air performance above 1200mA/cm² being due to the structure's inability to retain sufficient water to maintain optimum hydration of the membrane, when operating at high current densities. As a consequence, the resistance of the membrane increases with increasing current 25 density giving rise to a commensurate loss in overall performance. This is also seen in the fall-off in oxygen performance, with increasing current density at a similar point to the air operation (the use of oxygen by removing the mass transport issues seen with air operation confirms that the losses on air are not due to mass transport effects) as shown in Figures 1 and 2.

30

The structure of the non-woven gas diffusion substrate of the present invention, overcomes these problems of water retention and maintains membrane hydration out to

much higher current densities on both air and oxygen operation as can be seen from the cell potential versus current density plots for Example 1, 2 and 3 in Figures 1 and 2.

CLAIMS

1. A non-woven fibre web comprising a plurality of fibres in the x- and y-directions of average length greater than 5mm ("longer fibres") and a plurality of fibres of average length less than 3mm ("shorter fibres") wherein at least a proportion of the fibres of average length less than 3mm are orientated in the z-direction, and wherein the proportion of fibres of average length less than 3mm is at least 20% of the total weight of fibres, and wherein the density of the non-woven fibre web is from 0.1g/cm³ to 0.35g/cm³.

10

2. A non-woven fibre web comprising up to and including 80% by weight of fibres in the x- and y-directions of average length greater than 5mm ("longer fibres"), and 20% or more by weight of fibres of average length less than 3mm ("shorter fibres") wherein at least a proportion of the fibres of average length of less than 3mm are orientated in the z-direction, and wherein the density of the non-woven fibre web is from 0.1g/cm³ to 0.35g/cm³.

15

3. A non-woven fibre web according to claim 1 or claim 2 wherein the proportion of shorter fibres is suitably no more than 85% by weight of total fibres in the substrate.

20

4. A non-woven fibre web according to claim 3 wherein the proportion of shorter fibres is suitably no more than 70% by weight of the total fibres.

25

5. A non-woven fibre web according to any of claims 1 to 4 wherein the longer fibres suitably have a maximum average length of 50mm.

6. A non-woven fibre web according to claim 5 wherein the longer fibres are of average length of 5mm to 30mm.

30

7. A non-woven fibre web according to any preceding claim wherein the shorter fibres are of average length less than 2mm.

8. A non-woven fibre web according to claim 7 wherein the shorter fibres are of average length less than 1mm.

9. A non-woven fibre web according to any preceding claim wherein the shorter fibres have an average minimum length of 50 μ m.

10. A non-woven fibre web according to any preceding claim wherein the longer fibres and shorter fibres are independently selected from the group consisting of carbon, glass, silica, polymer, metal and ceramic fibres.

11. A non-woven fibre web according to claim 10 wherein the longer fibres and shorter fibres are carbon.

12. A non-woven fibre web according to any preceding claim wherein the density of the non-woven fibre web is from 0.1g/cm³ to 0.2g/cm³.

13. A process for the preparation of a non-woven fibre web according to any preceding claim, said process comprising the steps of:

- (i) dispersing the longer and shorter fibres in solution to form a slurry;
- (ii) adding one or more polymers (the "first polymer") to the slurry;
- (iii) draining the liquid from the slurry to form a web, or forming a continuous structure by the controlled deposition of the slurry onto a moving bed mesh;
- (iv) drying the web;
- (v) optionally placing the web in a solution of polymer (the "final polymer");
- (vi) drying the web; and
- (vii) heat treating the web.

14. A gas diffusion substrate comprising a non-woven fibre web as claimed in any one of claims 1 to 12, and a filler material.

15. A gas diffusion substrate according to claim 14 wherein the filler material comprises particulate carbon and a polymer.

16. A gas diffusion substrate according to claim 14 or claim 15 wherein the filler material comprises a catalyst material.

17. A gas diffusion electrode comprising a gas diffusion substrate as claimed in any 5 one of claims 14, 15 and 16, and an electrocatalyst material.

18. A gas diffusion electrode according to claim 17, wherein the electrocatalyst material comprises one or more electrocatalytic components and a polymer.

10 19. A gas diffusion electrode according to claim 18, wherein the electrocatalyst material is a precious metal or a transition metal as the metal or metal oxide, either unsupported or supported in a dispersed form on a carbon support; a carbon or an organic complex, in the form of a high surface area finely divided powder or fibre, or a combination of these options.

15 20. A membrane electrode assembly comprising a gas diffusion electrode as claimed in any one of claims 17, 18 and 19, a second gas diffusion electrode which may or may not be an electrode as claimed in any one of claims 17, 18 and 19, and a solid polymer membrane.

20 21. A membrane electrode assembly comprising a gas diffusion electrode as claimed in any one of claims 17, 18 and 19, a gas diffusion substrate which may or may not be a substrate as claimed in any one of claims 14, 15 and 16, and a solid polymer membrane, wherein an electrocatalyst layer is applied to the side of the membrane facing the gas diffusion substrate.

25 22. A membrane electrode assembly comprising a gas diffusion substrate as claimed in any one of claims 14, 15 and 16, a gas diffusion electrode which may or may not be an electrode as claimed in any one of claims 17, 18 and 19, and a solid polymer membrane, 30 wherein an electrocatalyst layer is applied to the side of the membrane facing the gas diffusion substrate.

23. A membrane electrode assembly comprising a gas diffusion substrate as claimed in any one of 14, 15 and 16, and a second gas diffusion substrate which may or may not be a substrate as claimed in any one of claims 14, 15 and 16, and a solid polymer membrane, wherein an electrocatalyst layer is applied to both sides of the solid polymer membrane.

24. A fuel cell comprising a gas diffusion substrate as claimed in any one of claims 14, 15 and 16.

10 25. A fuel cell comprising a gas diffusion electrode as claimed in any one of claims
17, 18 and 19.

卷之三

Fig. 1. Cell potential versus current density - air operation

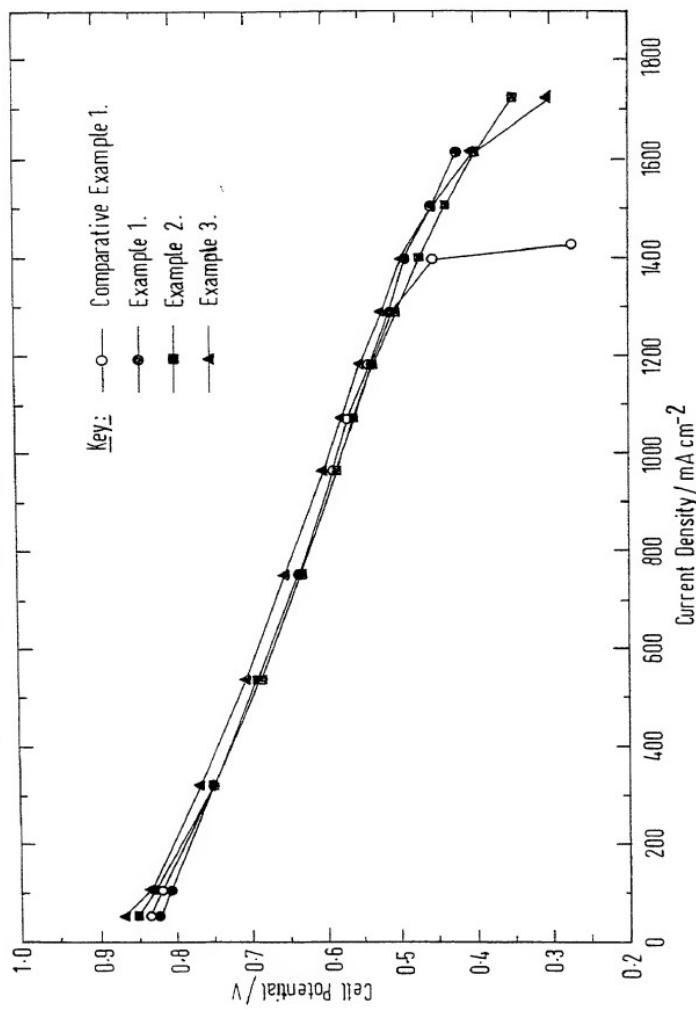
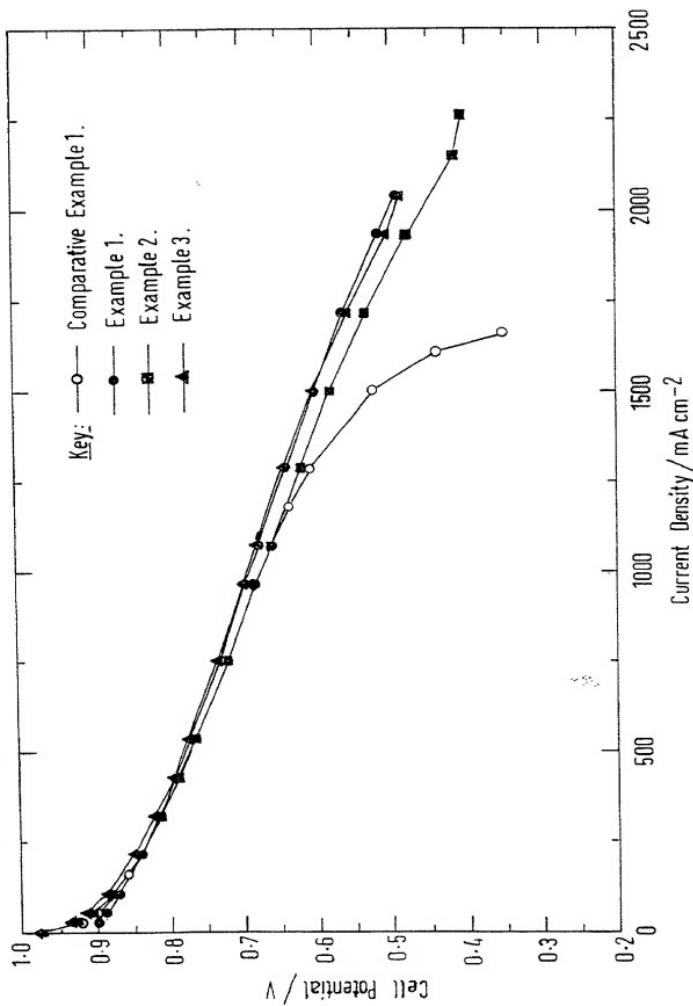


Fig.2. Cell potential versus current density -oxygen operation



JMYT-246US

Page 1 of 3

Declaration and Power of Attorney For Patent Application

English Language Declaration

As a below named Inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled

NONWOVEN WEB

The specification of which is attached hereto unless the following box is checked:

was filed on 09 February 2000 as

United States Application Number or PCT International Application Number PCT/CB00/00390, and was amended by a Preliminary Amendment filed along with the U.S. National Phase application.

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to patentability as defined in 37 CFR § 1.56.

I hereby claim foreign priority benefits under 35 U.S.C. §119(a)-(d) or § 365(b) of any foreign application(s) for patent or inventor's certificate, or § 365(a) of any PCT International application which designated at least one country other than the United States, listed below and have also identified below by checking the box, any foreign application for patent or inventor's certificate, or PCT International application having a filing date before that of the application on which priority is claimed:

Prior Foreign Application(s)

Priority Not Claimed

9903320.1

Great Britain

15 February 1999

(Number)

(Country)

(Day/Month/Year Filed)

I hereby claim the benefit under 35 U.S.C. § 119(e) of any United States provisional application(s) listed below.

I hereby claim the benefit under 35 U.S.C. § 120 of any United States application(s), or 365(c) of any PCT International application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International application in the manner provided by the first paragraph of 35 U.S.C. § 112, I acknowledge the duty to disclose information which is material to patentability as defined in 37 CFR § 1.56 which became available between the filing date of the prior application and the national or PCT international filing date of this application:

JMYT-246US

Page 2 of 3

(Application Number)

(Filing Date)

(Status - patented, pending, abandoned)

(Application Number)

(Filing Date)

(Status - patented, pending, abandoned)

POWER OF ATTORNEY: As a named Inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and transact all business in the Patent and Trademark Office connected therewith:

Robert L. Andersen

Reg. No. 25,771

Daniel N. Calder

Reg. No. 37,454

Justina L. Cohen

Reg. No. 39,140

Jacques L. Elkovicz

Reg. No. 21,738

Jack J. Jankevitz

Reg. No. 42,690

Christopher R. Lewis

Reg. No. 36,201

Larry B. Morris

Reg. No. 32,546

Kenneth N. Nigon

Reg. No. 81,546

Allan Rabkin

Reg. No. 19,112

Comille Jolly-Tornetta

Reg. No. 45,592

Lawrence F. Ashay

Reg. No. 24,515

Steven Bach

Reg. No. 46,530

Lowell L. Carson

Reg. No. 45,545

Kevin R. Casey

Reg. No. 32,117

Matthew I. Cohen

Reg. No. 45,133

Rey A. Connolly, IV

Reg. No. 41,719

Kevin W. Goldstein

Reg. No. 39,008

William P. House

Reg. No. 26,277

Scott A. McNeown

Reg. No. 25,028

Benjamin E. Leach

Reg. No. 33,612

Costas G. Krikalis

Reg. No. 42,565

Bruce M. Monroe

Reg. No. 33,602

Andrew J. Ney

Reg. No. 20,300

Steven Ng

Reg. No. 42,092

Pamela D. Telleris

Reg. No. 47,685

Paul F. Prestia

Reg. No. 23,031

James C. Simmons

Reg. No. 25,842

Jonathan H. Spadt

Reg. No. 45,122

Address all correspondence to: Christopher R. Lewis

Ratner & Prestia, Suite 301, One Westlakes, Bala Cynwyd, PA 19482-0980

Address all telephone calls to: Christopher R. Lewis at (610) 407-0700.

I hereby declare, that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Full name of sole or first inventor (given name, family name) John Malcolm Gascoyne

Inventor's signature

Date 14/8/01

Residence Bucks, United Kingdom

Citizenship British

Post Office Address Lynduska, Brails Green, Plecklow Ridge, High Wycombe

Bucks HP14 4BB, United Kingdom

Full name of second joint inventor, if any (given name, family name) Thomas Robertson Ralph

Second Inventor's signature

Date 14/8/01

Residence Reading, United Kingdom

Citizenship British

Post Office Address 91 Shirebury Road

Reading RG20 2QJ, United Kingdom

 Additional inventors are being named on separately numbered sheets attached hereto.

JMYT-246US

Page 3 of 3

300
Full name of third joint inventor, if any (given name, family name) Karen Legane Brown

Third inventor's signature [Signature] Date 14/8/01

Residence Reading, United Kingdom

Citizenship British

Post Office Address 45 Churchill Crescent, Sonning Common

Reading RG4 9PU, United Kingdom

Full name of fourth joint inventor, if any (given name, family name) _____

Fourth inventor's signature _____ Date _____

Residence _____

Citizenship _____

Post Office Address _____

Full name of fifth joint inventor, if any (given name, family name) _____

Fifth inventor's signature _____ Date _____

Residence _____

Citizenship _____

Post Office Address _____

Full name of sixth joint inventor, if any (given name, family name) _____

Sixth inventor's signature _____ Date _____

Residence _____

Citizenship _____

Post Office Address _____

Full name of seventh joint inventor, if any (given name, family name)

Seventh inventor's signature _____ Date _____

Residence _____

Citizenship _____

Post Office Address _____